Supplementary information

High-Performance van der Waals stacked transistors based on ultrathin $GaPS_4$ dielectrics

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1. XRD spectra of $GaPS_4$ powders.

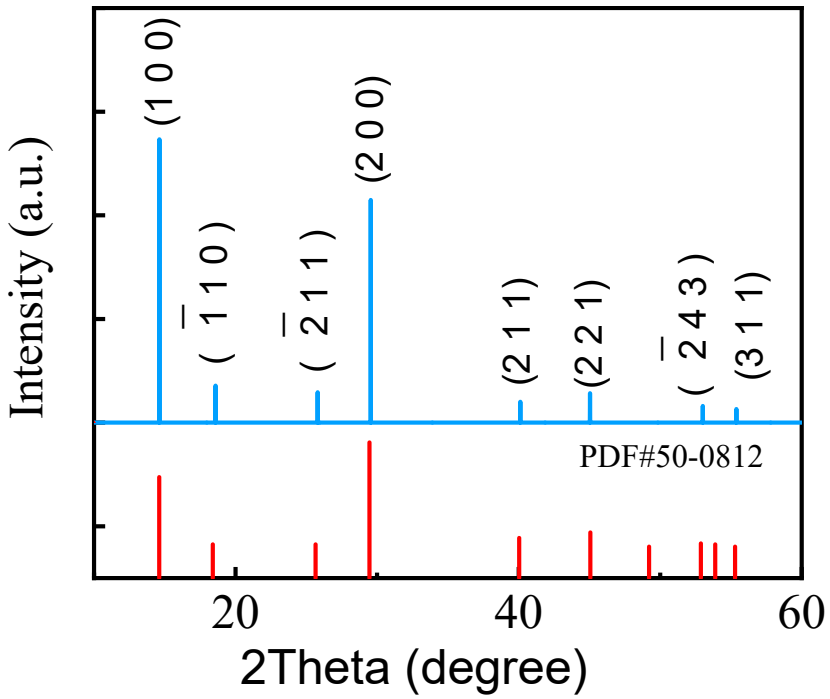
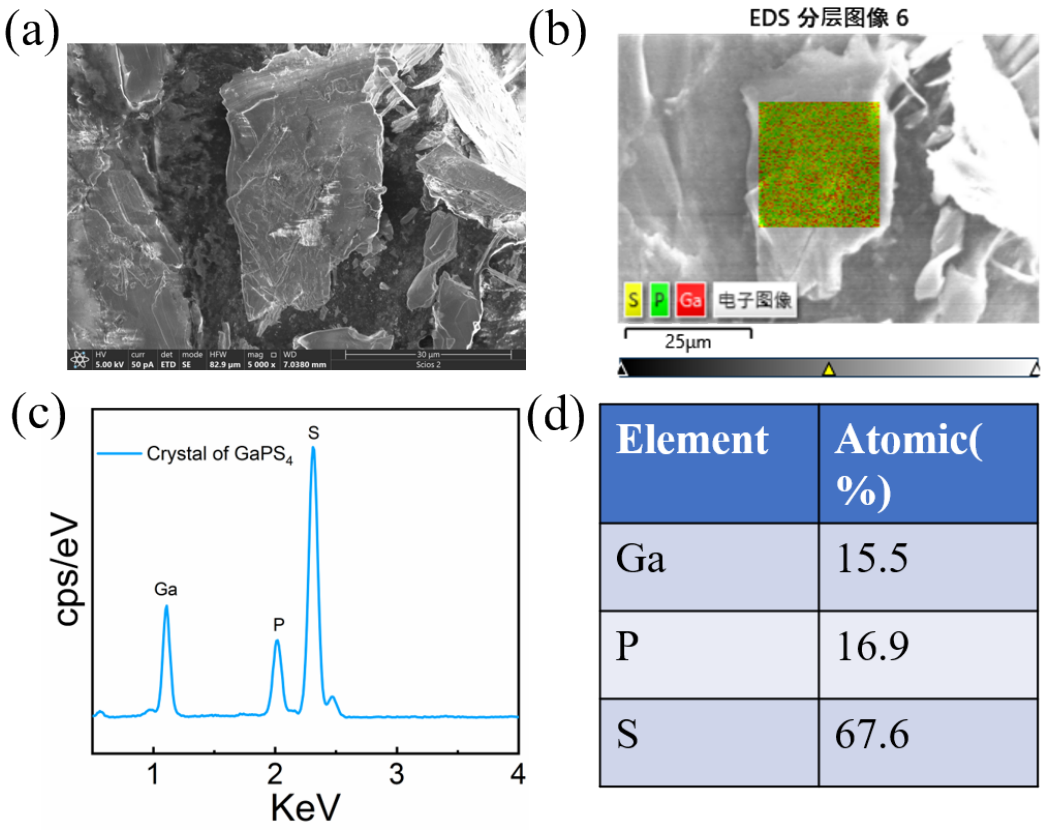


Fig. S1. XRD spectra for $GaPS_4$ powders in the blue line, and in comparison with the standard PDF card in the red line. The card number is (PDF#50-8012)

2. We use EDS to analyze the quality of GaPS4 single crystal grown by CVT. As shown in the figure, we can see that the proportion of Ga, P, and S elements is close to 1:1:4, indicating that the quality of our singer crystal is very well.



Element	Atomic(%)
Ga	15.5
P	16.9
S	67.6

Fig. S2. EDS spectrum of $GaPS_4$

3. Ultraviolet absorption Spectra of $GaPS_4$ thin Films with different thicknesses.

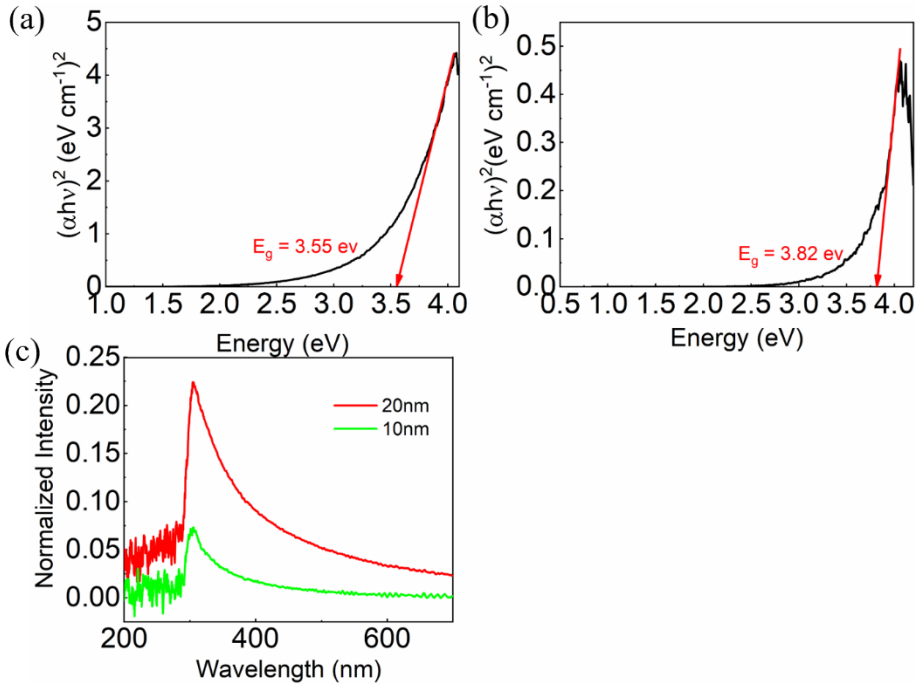


Fig. S3. (a) and (b) The fittings to these spectra to extract the optical band gap. (c) Absorption spectra of $GaPS_4$ flakes with different thicknesses.

4. UPS measurement of $GaPS_4$ and band misalignment between $GaPS_4$ and MoS_2.

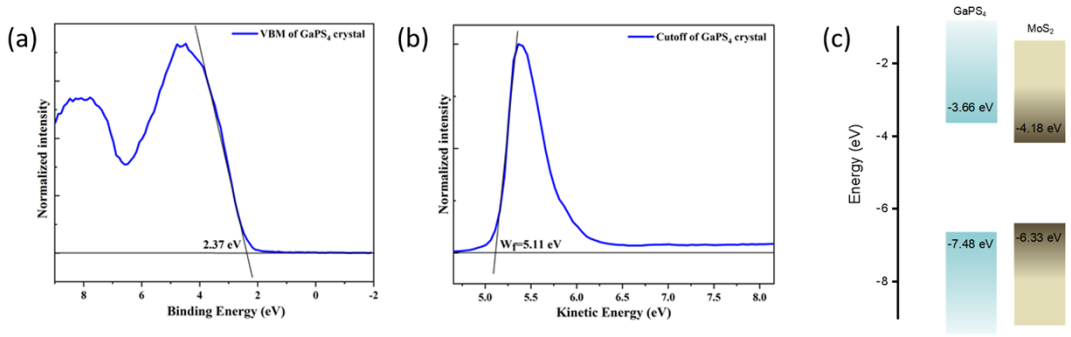


Fig. S4 (a) VBM of $GaPS_4$ crystal. (b) Work function of $GaPS_4$ crystal. (c) Band alignment between $GaPS_4$ and few-layer MoS_2.

5. The performance of different $GaPS_4$ gated MoS_2 FETs.

To investigate the thickness-dependent effect, we fabricated four $MoS_2/GaPS_4$ transistors with different thicknesses. Then, we display the device with better performance in Figure 3, and the characterization of other additional devices is shown in Figure S4 of the supplementary information. The comparison of the extracted parameters is shown in Table S1. The results we obtained show that FETs assembled from $MoS_2/GaPS_4$ with different thicknesses have similar gate responses, and the mobility and threshold voltage of different devices are different. This may be due to the influence of electrode contacts such as metal (Ti/Ni) or the quality of MoS_2 channels. Therefore, the thickness effect is not an important factor affecting device performance.

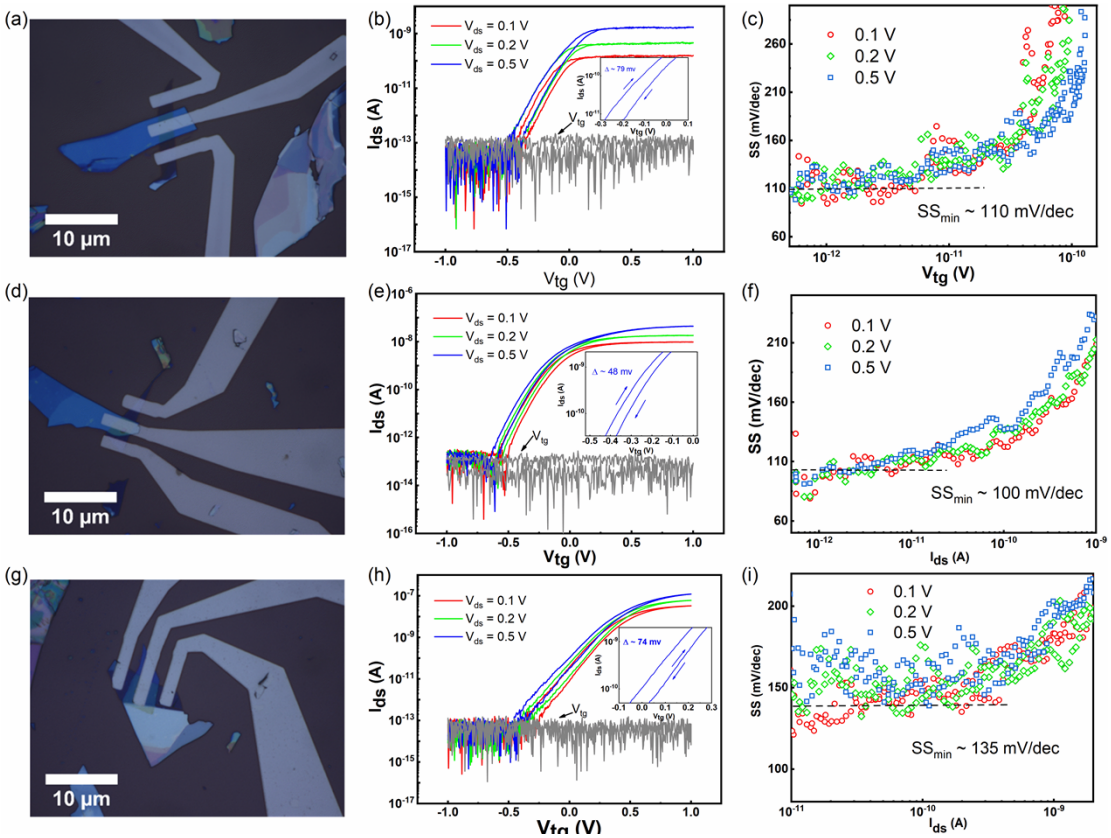


Fig. S5 Optical images, gate transfer curves, and SS responses for an additional three $GaPS_4$ gated MoS_2 FET transistors. (a)-(c) Optical image, gate transfer curves, and SS responses for device 2. (d)-(f) Optical image, gate transfer curves, and SS responses for device 3. (g)-(i) Optical image, gate transfer curves and SS responses for device 4.

6. The performance of $GaPS_4$ gated MoS_2 FETs.

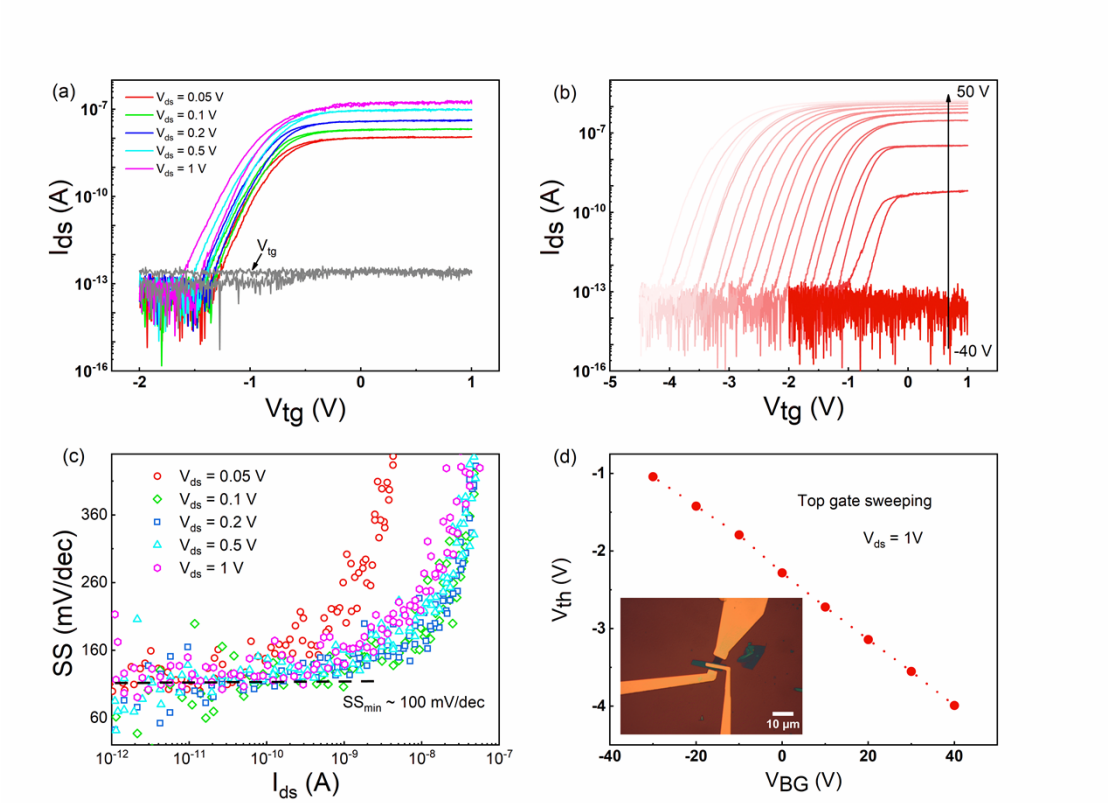


Fig. S6 The performance of FET (a) Typical dual-sweep transfer curves of the MoS_2/ $GaPS_4$ measured under different V_{ds} from 0.05 to 1 V. (b) Dual gated transfer curves of the MoS_2/ $GaPS_4$ FET under different back-gate voltages. (c) Extracted SS value versus I_{ds} characteristics of the device in a, showing a low SS value (< 70 mV/decade) for a wide I_{ds} range. (d) The extracted threshold voltage V_{th} from b is a function of V_{BG}.

Table S1. Extracted parameters for the performance of MoS_2 FETs using $GaPS_4$ as a top gate dielectric.

Device No.	Thickness of MoS_2 (nm)	Thickness of $GaPS_4$ (nm)	On/Off Ratio	Mobility ($cm^2/V{\cdot}s$)	Threshold voltage (V)	SS (mv/dec)
1(Figure 3)	7	22	10^7	14.2	-0.78	80
2	10	48	10^4	16.67	-0.46	100
3	4	20	10^5	8.78	-0.59	110
4	9	30	10^6	14.68	-0.27	135

7. Dielectric characteristics of $GaPS_4$

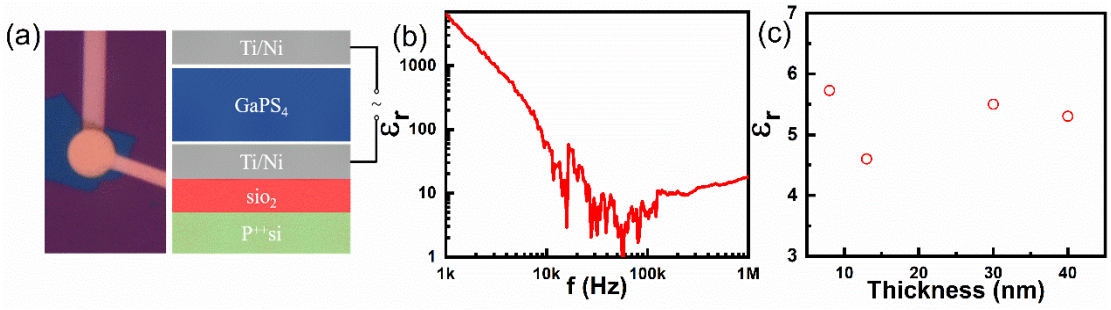


Fig. S7 (a) Optical image and schematic diagram of micro-sized $GaPS_4$ capacitor used for dielectric measurements, with a metal/$GaPS_4$/metal sandwich structure. (b) Extracted dielectric constant as a function of frequency from 1 kHz to 1000 kHz for a 40 nm thick flake. **(c)** Dielectric constant measured at 40 kHz for different thicknesses of flakes.

Table S2. Comparison of the dielectric properties and the corresponding FET performance of $GaPS_4$ with other 2D dielectric materials

Dielectric	**E_g**	**ε_r**	**FET channel**	**I_{on}/I_{off}**	**SS**	**refs**
SnP_2S_6	2.23	23	MoS_2	10^7	69.4	1
CaF_2	12.1	8.4	MoS_2	10^6	91	2
h-BN	6	3.9	MoS_2	10^6	80	3
SiP_2	2.14	8.1	MoS_2	10^5	-	4
$GaInS_3$	2.91	12	MoS_2	10^7	71.2	5
LaOBr	5.3	8	MoS_2	10^8	85	6
$MnAl_2S_4$	3.65	6.1	MoS_2	10^7	80	7
$Sr_2Nb_3O_{10}$	3.8	24.6	MoS_2	10^6	88	8
$In_2P_3S_9$	3.5	24	MoS_2	10^5	88	9
$GaPS_4$	3.55	5.3	MoS_2	10^7	80	This work

Reference

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